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2 **toxic pollutants for life-cycle impact assessment**

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Geographical scenario uncertainty in generic fate and exposure factors of toxic pollutants for life-cycle impact assessment

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Abstract

29

30 In environmental life-cycle assessments (LCA), fate and exposure factors account for the general

31 fate and exposure properties of chemicals under generic environmental conditions by means of

32 'evaluative' multi-media fate and exposure box models. To assess the effect of using different

33 generic environmental conditions, fate and exposure factorss of chemicals emitted under typical

34 conditions of (1) Western Europe, (2) Australia and (3) the United States of America were

35 compared with the multi-media fate and exposure box model USES-LCA. Comparing the results

36 of the three evaluative environments, it was found that the uncertainty in fate and exposure

37 factors for ecosystems and humans due to choice of an evaluative environment, as represented by

38 the ratio of the 97.5^{th} and 50th percentile, is between a factor 2 and 10. Particularly, fate and

39 exposure factors of emissions causing effects in fresh water ecosystems and effects on human

40 health have relatively high uncertainty. This uncertainty is mainly caused by the continental

41 difference in the average soil erosion rate, the dimensions of the fresh water and agricultural soil

42 compartment, and the fraction of drinking water coming from ground water.

43

44 **Keywords:** LCA; fate and exposure factors; ecotoxicity; human toxicity; evaluative environment.

45

46

1   Introduction

Characterisation factors for toxic pollutants, also called toxicity potentials, are substance-specific, quantitative representations of potential impacts per unit emission of a toxic substance. In environmental life cycle assessments of products (LCAs), these potentials are used to determine the relative contribution of a substance to toxicity related impact categories, such as human toxicity. Generally, generic characterisation factors are used for the comparison of toxic compounds in LCA case studies. These generic characterisation factors account for the general properties of the chemical, such as its toxicity and persistence under generic conditions by means of 'evaluative' multi-media fate and exposure box models (e.g. Hertwich *et al.*, 2001, 2002).

Before generic characterisation factors can be calculated, the choices of the landscape and human characteristics have to be discussed. In the calculation of current toxicity potentials, 'evaluative' environments applied represent in most cases the continental scale, such as Western Europe or the United States of America (Huijbregts *et al.* 2000a; Hertwich *et al.*, 2001). To assess the uncertainty on generic toxicity potentials of using different 'evaluative' environments, fate and exposure characteristics of toxic substances emitted under typical conditions of (1) Western Europe, (2) Australia and (3) the 48 contiguous States of the United States of America will be compared with the 'evaluative' multi-media fate and exposure box model USES-LCA, originally applied by Huijbregts *et al.* (2000a) for West European conditions. The present article briefly outlines the multi-media fate and exposure part of USES-LCA and the conversion of the model for Australia and the United States. For the three evaluative environments, fate and exposure factors for humans and ecosystems of 375 substances emitted to four different compartments are compared and the uncertainties found are discussed.

2 Method

2.1 Fate and exposure factors

USES-LCA, based on the Uniform System for the Evaluation of Substances 2.0 (USES 2.0) as developed by RIVM *et al.* (1998), is designed to calculate the potential impacts of toxic substances emitted in Western Europe on humans and ecosystems. USES-LCA models fate, exposure and effects for a time horizon of 20, 100, and 500 years and an infinite time horizon (Huijbregts *et al.*, 2000a, 2001). The fate and exposure part of USES-LCA are briefly explained below. Huijbregts *et al.* (2000a) give a more extensive elaboration of the model.

The fate module of USES-LCA consists of the nested multi-media fate model Simplebox 2.0 (Brandes *et al.*, 1996), which is used to calculate environmental concentrations in the various environmental media. "Nested" means that chemicals can be transported from one scale to a higher scale and vice versa. USES-LCA has two spatial scales (continental and hemispheric) and three climate zones, reflecting arctic, moderate and tropic climatic zones of the Northern hemisphere (Figure 1). The continental scale is defined within the moderate climate zone and consists of six compartments: air, fresh water, seawater, natural soil, agricultural soil, and industrial soil. The hemispheric scale comprises the arctic, moderate and tropical climate zones. Every climatic zone is divided in three compartments: air, seawater and soil. Because the hemispheric scale is modelled as a closed system without transport across the system boundaries, emitted substances cannot escape.

<Figure 1 about here>

96   The exposure module is used to calculate the change in exposure for the human population via

97   air, drinking water and food products. For an ecosystem, the change in exposure equals the

98   change in the dissolved environmental concentration predicted by the fate module. In formula

99   this means that

100    $$F_{x,i \to j} = \sum_s N_{j,s} \times \frac{dD_{x,j,s}}{dM_{x,i}}$$ (1)

101   in which $F_{x,i \to j}$ represents the fate and exposure factor that accounts for transport of substance x

102   from compartment i to target j (dimensionless for humans or days for ecosystems), $N_{j,s}$ is the total

103   number of target j at scale s (in number of individuals for humans or in m^3 of water, sediment or

104   soil for ecosystems) and $dD_{x,j,s}/dM_{x,i}$ is the marginal change in exposure of substance x by target j

105   at scale s ($dD_{x,j,s}$ in kg.day^{-1}. person^{-1} for humans or in kg.m^{-3} for ecosystems) caused by a

106   marginal change in the emission of substance x to compartment i ($dM_{x,i}$ in kg.day^{-1}).

107

108   *2.2   Continental scenarios*

109

110   The following geographical differences between Western Europe, Australia and the United States

111   were taken into account (Table 1):

112   • Environmental parameters and human exposure characteristics on the continental scale were

113   set for conditions representative for the continents under consideration;

114   • Australia was nested in the tropical zone, while Western Europe and the United States were

115   nested in the moderate zone of the global scale; and

116   • For the Australian calculations, environmental parameters on the three climate zones, such as

117   land-sea ratio, were set for the Southern hemisphere, while for Western Europe and the

118   United States environmental parameters on the three climate zones were set for the Northern

119   hemisphere.

6

120

121   <Table 1 about here>

122

123   *2.3   Calculations*

124

125   The infinite time horizon setting was used in all the USES-LCA calculations. For each

126   continental scenario, fate and exposure factors of 375 substances were calculated for the

127   environmental endpoints (1) fresh water aquatic ecosystems, (2) marine aquatic ecosystems, (3)

128   terrestrial ecosystems, and (4) humans after emission to respectively air, fresh water, seawater

129   and agricultural soil. Sediment ecosystems and emissions to industrial soil were not included in

130   the analysis, because Huijbregts (2001) found that fate and exposure factors for sediment and

131   aquatic ecosystems, and fate and exposure factors after emissions to industrial soil and

132   agricultural soil are highly correlated. These high correlations imply that the same type of

133   geographical differences will be found for sediment and aquatic ecosystems and emissions to the

134   two soil compartments, respectively. For humans, not only full fate and exposure factors, taking

135   into account all exposure pathways, were calculated, but also separate fate and exposure factors

136   for the pollutant exposure routes (a) food; (b) drinking water, and (c) inhalation. The datasets of

137   Huijbregts *et al.* (2000a) and Hertwich *et al.* (2001) were used to specify substance-specific

138   parameters.

139

140   For each human health and ecological endpoint, the fate and exposure factors of all 375

141   substances for all four emission compartments were used to derive linear regression equations of

142   the form

143

7

144 $$\log F_{j,Sn} = a \times \log F_{j,Sm} + b \tag{2}$$

145

146 in which S_n and S_m are continental scenarios. We optimized the regression equations using a

147 linear least squares fit to find appropriate values of the slope (a) and intercept (b) of the

148 regressions. Apart from the regression parameters a and b, the correlation coefficient (r^2), the

149 residual standard error (SE), and the uncertainty factor k were derived. The correlation coefficient

150 (r^2) represents the fraction of explained variance by the regression equation, while the residual

151 standard error (SE) represents the average residual error between the calculated F with USES-

152 LCA and the estimated F with the regression equation. The SE can be converted to an

153 uncertainty factor k which is defined such that 95% of the values of a stochastic variable are

154 within a factor k from the median of a lognormal distribution (Slob, 1994). The uncertainty factor

155 k can be calculated from SE by

156

157 $$k = \sqrt{\frac{97.5p}{2.5p}} = 10^{1.96 \times SE} \tag{3}$$

158

159 in which $2.5p$ and 97.5p are respectively the 2.5$^{\text{th}}$ percentile and 97.5$^{\text{th}}$ percentile of the

160 uncertainty distribution..

161

3   Results

Table 2 shows the statistics of the regression equations for the fate and exposure factors of the three continental scenarios. For all ecosystem fate and exposure regression equations it can be seen that the explained variance r^2 is high (> 0.95), while the uncertainty factor k is always lower than 5, except for the fresh water fate and exposure factors (FwF) of the US versus Australia with a uncertainty factor k of 8. As the regression coefficient a approaches unity for all ecosystem regression equations, the relationship between the ecosystem fate and exposure factors of the three continental scenarios is approximately linear. Systematic differences between the ecosystem fate and exposure factors of the three continental scenarios, represented by the factor 10^b, remain within a factor 3, except for the comparison of Australian FwFs with the two other continental FwFs (a factor of 10).

For the human fate and exposure regression equations it can also be seen in Table 2 that the explained variance r^2 is high (> 0.90). The uncertainty factor k in the human fate and exposure regression equations can be up to a factor of 25. The uncertainty in inhalation exposure regression equations do not largely contribute to the uncertainty in total human fate and exposure factors (HuF), in contrast to the uncertainty in exposure via food and drinking water intake. Again, the regression coefficient a approaches unity for the human fate and exposure regression equations, resulting in an approximate linear relationship between the HuFs of the three continental scenarios. Systematic differences between the Australian HuFs and the HuFs of the two other continents are relatively high (up to a factor of 70).

<Table 2 about here>

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4   Discussion

4.1   Evaluative environment

The uncertainty in fate and exposure factors for ecosystems and humans due to choice of an

evaluative environment, represented by the uncertainty factor k, is between a factor 2 and 10.

Previous investigations indicated that uncertainty in chemical-specific parameters, such as

degradation rates, lead to uncertainty factors of 4 to 50 for human fate and exposure factors

(Huijbregts et al., 2000b, Hertwich et al., 1999, 2000) and uncertainty factors of 2 to 500 for

ecosystem fate and exposure factors (Huijbregts et al., 2000b), depending on the chemical and

emission compartment considered. Comparing the influence of the choice of an evaluative

environment and parameter uncertainty on fate and exposure factors, our results indicate that the

influence of the choice of an arbitrary evaluative environment may only be relevant for the fresh

water aquatic and human fate and exposure factors.

Uncertainty in the regression equations for fresh water aquatic and human fate and exposure

factors can be clarified by the fact that the three evaluative environments do not change fate and

exposure in the same way (1) for all substances and (2) for all initial emission compartments. A

combination of the following factors cause substance-specific differences in the fate and

exposure factors of the three evaluative environments:

- The lower Australian fresh water area compared to Western Europe and the United States

    results in lower chemical *residence times* in the fresh water compartment and thus lower

    FwFs, if burial of sediment or advection to the marine environment are the most important

    chemical removal processes from the fresh water compartment. The lower fresh water area

    also results in a higher dissolved *concentration* of chemicals in the fresh water compartment

10

211   and thus higher HuFs for drinking water and fish intake, if degradation or volatilisation are the

212   most important chemical removal processes from the fresh water compartment. Differences

213   are up to a factor of 6;

214   • The lower depth of the Australian fresh water compartment compared to Western Europe and

215   the United States results in lower chemical *residence times* and thus lower FwFs, if burial of

216   sediment, volatilisation or advection to the marine environment are the most important

217   removal processes for chemicals from the fresh water compartment. The lower depth also

218   results in a higher dissolved *concentration* of chemicals and thus higher HuFs for drinking

219   water and fish intake, if degradation is the most important chemical removal processes from

220   the fresh water compartment. Differences are up to factor of 5;

221   • The lower rainrate and fraction of precipitation that runs off soil in Australia compared to

222   Western Europe and the United States result in a smaller transfer from soil to fresh water of

223   substances with relatively low solid-water partition coefficients. Australian FwFs and HuFs

224   for drinking water and fish intake of these of substances are up to factor of 4 lower; and

225   • The relatively high soil erosion rate in Australia results in a higher average net sedimentation

226   and consequently a higher burial of sediment in the Australian fresh water compartment

227   compared to Western Europe and the United States. As sedimentation is a significant removal

228   route for water persistent, hydrophobic, low-volatility substances, and metals with relatively

229   high solid-water partition coefficients, both the residence time and concentration will be lower

230   for these type of substances in Australian surface waters, resulting in lower FwFs and HuFs

231   for drinking water and fish intake of these of substances (up to a factor of 7).

232

233   The following differences in the evaluative environments cause emission compartment-specific

234   deviations in the fate and exposure factors:

11

235   •   The lower Australian fresh water area compared to Western Europe and the United States

236   results in a smaller chemical transfer from air to fresh water, lowering the Australian FwFs

237   and HuFs of substances emitted to air, seawater and agricultural soil (up to a factor 5), while

238   the smaller chemical transfer from air to fresh water does not change the FwFs and HuFs of

239   substances emitted to fresh water;

240   •   The lower rainrate and fraction of precipitation that runs off soil in Australia compared to

241   Western Europe and the United States results in smaller chemical transfer from soil to the

242   fresh water, lowering the Australian FwFs and HuFs of substances emitted to agricultural soil,

243   are and seawater (up to factor of 4). The smaller chemical transfer from soil to fresh water

244   does not change the FwFs and HuFs of substances emitted to fresh water; and

245   •   The Australian HuFs of substances emitted to agricultural soils are on average a factor of 160

246   lower compared to the Western European HuFs, while substances emitted to air, fresh water

247   and seawater are on average a factor of 20 lower. The combination of the larger area of

248   agricultural soil, the higher erosion rate and the smaller fraction of drinking water coming

249   from ground water in Australia compared to Western Europe explain the lower Australian

250   HuFs of substances emitted to agricultural soils. The larger area of agricultural soil and the

251   higher erosion rate in Australia result in lower soil and ground water concentrations per unit

252   emission and, as a result, lower concentrations in agricultural food products and drinking

253   water coming from ground water. These differences lower the Australian HuFs of substances

254   with agricultural food products and drinking water as dominant human exposure routes, which

255   is the case for most substances emitted to the agricultural soil.

256

257

4.2   Site dependency

A number of studies addressed the potential importance of spatial differention in the impact assessment of toxic substances in LCA (Hertwich *et al.* ,2000; Krewitt *et al.* 2001; McKone *et al.*, 2000; Nigge, 2000; Potting, 2000; Schulze, 2001). In the context of spatial differentiation, the present study shows systematic differences between continental FwFs and HuFs of Australia versus Western Europe and the United States between a factor of 8-16 and 9-45, respectively. These systematic differences in continental fate and exposure factors are caused by differences in, for instance, human population numbers, temperature and hydroxyl radical concentration at the different continents. The significance of these systematic continental differences will depend, however, on (1) the spatial range of the pollutant and (2) the spatial scale at which important regional parameters vary (Hertwich *et al.*, 2002). According to Hertwich & McKone (2001), the spatial range of a pollutant is the mean distance that a pollutant travels once it is released and can be calculated by the product of persistence and average mobility. Following this definition, USES-LCA calculated that for about 90% of the substances the spatial range is smaller than the continental scales as used in our calculations. If the spatial scale at which important regional parameters vary is smaller than the continental scale, scenarios are required at a smaller geographical scale. The question whether scenarios are required at a smaller scale may be answered by comparing the state-to-state (or region-to-region) variability in fate and exposure characteristics *within* the three continents with the variation of fate and exposure characteristics *between* these three continents. Further research is required to reveal whether the systematic differences found between the three evaluative environments are of direct relevance for LCA purposes.

13

5   Conclusions

The model USES-LCA, suitable for Western European conditions, can be modified to reflect conditions of other geographical regions, such as Australia and the United States. For each of the three geographical regions, USES-LCA was applied to calculate fresh water, marine, terrestrial and human fate and exposure factors of 375 substances emitted to four different compartments. From the comparison of the three evaluative environments it was found that fate and exposure factors of emissions causing effects in fresh water ecosystems and human health effects have relatively high uncertainty. In this respect, the choice of the average soil erosion rate, the dimensions of the fresh water and agricultural soil compartment, and the fraction of drinking water coming from ground water is considered important in the calculation of generic fate and exposure factor for toxic pollutants.

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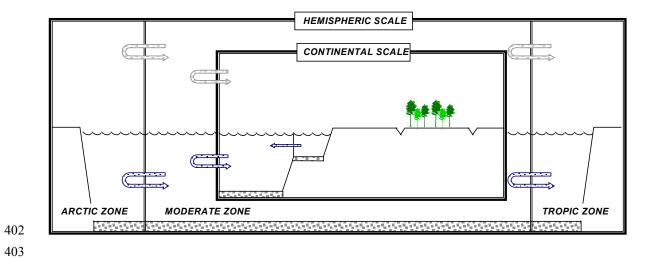390 **Table 1:** Input parameters for fate analysis and human exposure assessment

	Unit	Australia	US	W-Europe	Ref.
Fate analysis					
Fresh water area [C]	km^2	$6.0 \cdot 10^4$	$3.5 \cdot 10^5$	$1.1 \cdot 10^5$	a-c
Natural soil area [C]	km^2	$3.0.10^6$	$3.9 \cdot 10^6$	$2.1 \cdot 10^6$	b-d
Agricultural soil area [C]	km^2	$4.5.10^6$	$3.8 \cdot 10^6$	$9.6 \cdot 10^5$	b-e
Industrial/other soil area [C]	km^2	$8.0.10^4$	$3.3 \cdot 10^5$	$3.6 \cdot 10^5$	a-c, f
Sea area [C]	km^2	$2.5.10^6$	$1.6 \cdot 10^6$	$3.6 \cdot 10^6$	c, g, h
Suspended particles (fresh water) [C]	$kg.m^{-3}$	$3.0 \cdot 10^{-2}$	$3.0 \cdot 10^{-1}$	$2.5 \cdot 10^{-2}$	b, c, i, j
Settling velocity of suspended particles [C]	$m.d^{-1}$	10	13	2.5	b, c
Depth (fresh water) [C]	m	1	5	3	b, c, k
Temperature [C]	°C	22	11.6	12	b, c, l
Rainrate [C]	$mm.year^{-1}$	450	835	700	b, c, m
Soil erosion rate [C]	$mm.year^{-1}$	$2.0 \cdot 10^{-1}$	$5.0 \cdot 10^{-2}$	$3.0 \cdot 10^{-2}$	b, c, n
Wind speed [C]	$m.s^{-1}$	3.2	4.4	3.0	b, c, l
Fraction of precipitation that runs off soil [C]	-	0.12	0.27	0.25	b, c, o
Fraction land [M]	-	0.04	0.50	0.50	p
Fraction sea [M]	-	0.96	0.50	0.50	p
Fraction land [T]	-	0.21	0.30	0.30	p
Fraction sea [T]	-	0.79	0.70	0.70	p
Fraction land [A]	-	0.33	0.40	0.40	p
Fraction sea [A]	-	0.67	0.60	0.60	p
OH-radicals [C, M, T, A]	$mol.cm^{-3}$	$6.0 \cdot 10^5$	$5.0 \cdot 10^5$	$5.0 \cdot 10^5$	q
Human exposure assessment					
Fraction drinking water from surface water [C]	-	0.8	0.5	0.4	b, c, r
Fraction drinking water from ground water [C]	-	0.2	0.5	0.6	b, c, r
Daily intake of drinking water [C]	$l.day^{-1}$	1.8	1.4	1.4	c, s, t
Daily intake of fish [C]	$kg_{wwt}.day^{-1}$	0.03	0.02	0.03	c, s, u
Daily intake of leaf crops [C]	$kg_{wwt}.day^{-1}$	0.73	0.66	0.77	c, s, u
Daily intake of root crops	$kg_{wwt}.day^{-1}$	0.25	0.08	0.18	c, s, u
Daily intake of meat [C]	$kg_{wwt}.day^{-1}$	0.20	0.13	0.26	c, s, u
Daily intake of dairy products [C]	$kg_{wwt}.day^{-1}$	0.39	0.50	0.28	c, s, u
Number of humans [C]	-	$1.8 \cdot 10^7$	$2.6 \cdot 10^8$	$3.7 \cdot 10^8$	p
Number of humans [M]	-	$6.1 \cdot 10^7$	$2.5 \cdot 10^9$	$2.5 \cdot 10^9$	p
Number of humans [T]	-	$6.1 \cdot 10^8$	$2.3 \cdot 10^9$	$2.3 \cdot 10^9$	p
Number of humans [A]	-	$1.0 \cdot 10^4$	$9.9 \cdot 10^7$	$9.9 \cdot 10^7$	p

391 [C] = Continental scale, [M] = Moderate zone, [A] = Arctic zone, [T] = Tropical zone; [a] CWA (1980); [b] McKone *et al.*

392 (2000); [c] Huijbregts (1999); [d] ABS (2001); [e] NASS (2001); [f] US Census Bureau (2000); [g] Bowen *et al.* (1996); [h] NOAA

393 (2001); [i] VWRDW (2001); [j] DLWC (2001); [k] Roser (2001); [l] BMA (2001); [m] Beckman (1996); [n] Noble *et al.* (1996); [o]

394 Crabb (1997); [p] Anonymous (1997); [q] Atkinson (1988); [r] NLWRA (2001); [s] USEPA (1999); [t] ABS (1999); [u] ABS (1998).

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Table 2: Regression characteristics of the fate and exposure factors for the three continental scenarios

			Statistics		
Regression equation	a	b	r^2	SE	k
Fresh water aquatic ecosystems					
$\log(FwF_{EU}) = a.\log(FwF_{US}) + b$	1.02	-0.24	0.99	0.22	2.7
$\log(FwF_{AU}) = a.\log(FwF_{EU}) + b$	1.03	-0.96	0.98	0.37	5.3
$\log(FwF_{AU}) = a.\log(FwF_{US}) + b$	1.06	-1.21	0.97	0.47	8.4
Marine aquatic ecosystems					
$\log(MaF_{EU}) = a.\log(MaF_{US}) + b$	0.97	0.14	0.99	0.19	2.3
$\log(MaF_{AU}) = a.\log(MaF_{EU}) + b$	1.06	-0.49	0.99	0.30	3.8
$\log(MaF_{AU}) = a.\log(MaF_{US}) + b$	1.03	-0.34	0.98	0.37	5.3
Terrestrial ecosystems					
$\log(TeF_{EU}) = a.\log(TeF_{US}) + b$	1.01	-0.10	1.00	0.13	1.8
$\log(TeF_{AU}) = a.\log(TeF_{EU}) + b$	1.00	-0.28	0.99	0.25	3.1
$\log(TeF_{AU}) = a.\log(TeF_{US}) + b$	1.01	-0.38	0.99	0.30	3.9
Humans (total)					
$\log(HuF_{EU}) = a.\log(HuF_{US}) + b$	1.03	0.62	0.95	0.38	5.5
$\log(HuF_{AU}) = a.\log(HuF_{EU}) + b$	0.96	-1.63	0.91	0.52	10.3
$\log(HuF_{AU}) = a.\log(HuF_{US}) + b$	1.01	-0.90	0.91	0.52	10.7
Humans (food)					
$\log(HuF_{EU}) = a.\log(HuF_{US}) + b$	1.04	0.76	0.95	0.43	7.0
$\log(HuF_{AU}) = a.\log(HuF_{EU}) + b$	0.96	-1.78	0.95	0.42	6.8
$\log(HuF_{AU}) = a.\log(HuF_{US}) + b$	1.00	-0.96	0.93	0.50	9.6
Humans (drinking water)					
$\log(HuF_{EU}) = a.\log(HuF_{US}) + b$	1.04	0.79	0.99	0.28	3.5
$\log(HuF_{AU}) = a.\log(HuF_{EU}) + b$	0.97	-1.85	0.96	0.69	22.4
$\log(HuF_{AU}) = a.\log(HuF_{US}) + b$	1.01	-1.07	0.95	0.71	24.4
Humans (air)					
$\log(HuF_{EU}) = a.\log(HuF_{US}) + b$	0.99	0.20	1.00	0.16	2.1
$\log(HuF_{AU}) = a.\log(HuF_{EU}) + b$	0.99	-1.23	1.00	0.29	3.6
$\log(HuF_{AU}) = a.\log(HuF_{US}) + b$	0.98	-1.10	1.00	0.32	4.3

396 AU = Australia; US = United States of America; EU = Western Europe; FwF = Fresh water fate and

397 exposure factor; MaF = Marine fate and exposure factor; TeF = Terrestrial fate and exposure factor; HuF

398 = Human fate and exposure factor; r^2 = explained variance; SE = residual standard error; k = uncertainty

399 factor.

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HEMISPHERIC SCALE

CONTINENTAL SCALE

ARCTIC ZONE

MODERATE ZONE

TROPIC ZONE

402

403

403 **Figure 1:** Schematic representation of transportation routes between different compartments and

404 geographical scales after Brandes *et al* (1996)